

# MIXING OF CO<sub>2</sub> AND CH<sub>4</sub> IN GAS RESERVOIRS: CODE COMPARISON STUDIES

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## ABSTRACT

Simulation of the mixing of carbon dioxide and methane is critical to modeling gas reservoir processes induced by the injection of carbon dioxide. We have compared physical property estimates and simulation results of the mixing of carbon dioxide and methane gases from four numerical simulation codes. Test Problem 1 considers molecular diffusion in a one-dimensional stably stratified system. Test Problem 2 considers molecular diffusion and advection in an unstable two-dimensional system. In general, fair to good agreement was observed between the codes tested.

## INTRODUCTION

Depleted oil and gas reservoirs are promising targets for carbon sequestration by direct carbon dioxide (CO<sub>2</sub>) injection because of their available volume and proven integrity against leakage. The ability to simulate accurately the mixing of supercritical CO<sub>2</sub> and methane (CH<sub>4</sub>) is essential for modeling reservoir processes under injection by CO<sub>2</sub>. We have undertaken a comparison of reservoir simulators focused on two test problems to compare simulation capabilities applicable to carbon sequestration in oil and gas reservoirs as part of a broader code intercomparison study [1]. Because the results of reservoir simulation are strongly dependent on the real gas properties of the gas mixtures, the first part of the study involved comparison of densities, viscosities, and solubilities from the different simulators for the pure end-member gases (CO<sub>2</sub> and CH<sub>4</sub>) and 50% mole fraction mixtures. In order to compare modeling capabilities, we defined two test problems that engage key processes involved in CO<sub>2</sub>-CH<sub>4</sub> mixing (Figure 1). Problem 1 considers the mixing by molecular diffusion and advection of a stably stratified one-dimensional column 100 m in height with the light gas (CH<sub>4</sub>) on the top and the heavy gas (CO<sub>2</sub>) on the bottom. Mixing around the interface is mostly by molecular diffusion, although nonzero permeability allows minor advection to occur due to gas expansion upon mixing at the interface. Problem 2 considers the mixing by advection and diffusion of gases initially side by side in a vertical 100 m x 100 m reservoir. Gravity effects cause the dense CO<sub>2</sub> gas to flow downward while the lighter CH<sub>4</sub> migrates upward.

In this paper, we present comparisons of physical properties of gas mixtures as estimated by the various codes along with results from the four codes for the two test problems. The purpose of this paper is to show comparisons of simulated results to build confidence in the use of numerical simulation for carbon

sequestration studies, as well as to provide test problem specifications and results that can be used by researchers to test and verify their own simulation capabilities.

## DESCRIPTION OF CODES

Four numerical simulation codes have been used for physical property estimation and Test Problems 1 and 2. These are as follows: CHEMTOUGH, developed by Industrial Research Limited, New Zealand, a geochemical modeling extension of TOUGH2; GEM, a reservoir simulator developed by Computer Modelling Group (CMG), Canada; SIMUSCOPP, a reservoir simulator developed by Institut Français du Pétrole (IFP), France; and TOUGH2/EOS7C, a special gas module for the TOUGH2 reservoir simulator [2] developed by Lawrence Berkeley National Laboratory, USA. Each of these codes uses its own methods for calculating physical properties of gas mixtures. Only CHEMTOUGH and TOUGH2/EOS7C share a common heritage and thus calculate flow and transport by the same methods.

## COMPARISON OF PHYSICAL PROPERTY ESTIMATES

To the extent that physical properties strongly affect flow and transport, the first comparison we present is for density ( $\rho$ ), viscosity ( $\mu$ ), and solubility of  $\text{CO}_2$  and  $\text{CH}_4$  gas mixtures. Carbon dioxide undergoes large changes in density and viscosity as it passes through the critical region. The critical pressure and temperature of  $\text{CO}_2$  (73.8 bars, 31.0 °C) will be reached in the subsurface at depths greater than approximately 800 m. Thus  $\text{CO}_2$  will most commonly be supercritical in the subsurface. Therefore, we present estimates of physical properties at both subcritical (40 bars) and supercritical (100 bars) conditions. For brevity, we present in Tables 1 and 2 physical properties only for the end members and 50-50 mixtures. We have included reference values either from published data or from more detailed estimation methods as noted.

Examination of the Tables reveals some areas of general agreement as well as scatter in the estimates. Three of the codes estimate densities at 40 and 100 bars accurately, with the exception of pure  $\text{CO}_2$  at 100 bars, where all codes underestimate the value. The equation of state used by CHEMTOUGH is accurate for temperatures between 50–350 °C. The temperature of comparison (40 °C) is slightly outside of this range, and this is reflected in the poor estimate of  $\text{CO}_2$  density. Viscosities are reasonably well estimated at high pressures. At low pressures, one code (SIMUSCOPP), which uses the Lohrenz-Bray-Clark viscosity model, estimates a lower viscosity for pure  $\text{CO}_2$  than for pure  $\text{CH}_4$ . As for solubilities, given as mole fraction of the gas component in the aqueous phase ( $x_1^{\text{CH}_4}$ ,  $x_1^{\text{CO}_2}$ ), there is fair agreement at 40 bars. However, at 100 bars, two codes (CHEMTOUGH and TOUGH2/EOS7C) overestimate pure  $\text{CO}_2$  solubility because they use a Henry's Law formulation that does not consider corrections applicable at high pressures. In general, no single code is consistently more accurate than the others, and all of them give good to fair approximations of physical properties.

## TEST PROBLEM 1. MIXING OF STABLY STRATIFIED GASES

In this problem,  $\text{CO}_2$  and  $\text{CH}_4$  gases are placed in contact one on top of the other and allowed to mix as controlled by diffusion and associated flow at 40 bars, 40 °C. Mixing in the one-dimensional system is limited because the denser gas ( $\text{CO}_2$ ) is on the bottom and the lighter gas ( $\text{CH}_4$ ) is on the top. The domain, properties, boundary and initial conditions are shown in Figure 1. All of the boundaries are closed and the problem is considered isothermal. Although the problem is dominated by diffusion, small advective fluxes arise as diffusive mixing around the interface leads to density changes that affect gas pressure.

The results of Test Problem 1 are shown in Figure 2a by the gas mole fraction of  $\text{CO}_2$  ( $x_g^{\text{CO}_2}$ ), where  $x_g^{\text{CH}_4} = 1 - x_g^{\text{CO}_2}$  in this binary system. As shown in Figure 2a, the fundamental process of binary diffusive mixing is captured by all of the codes, with slight differences in diffusion rate. Note in Figure 2b that the pressure in the system increases, a result of the effect on gas density of the mixing between  $\text{CO}_2$  and  $\text{CH}_4$ . In direct relation to the overestimate of pure  $\text{CO}_2$  density (see Table 1) and the underestimate of gas mixture density by CHEMTOUGH (see Table 1), this code predicts larger pressure increases than any of the other codes. Variations in the results are likely due mostly to differences in physical property

estimates rather than to differences in modeling of the physical process of molecular diffusion and advection.

TABLE 1

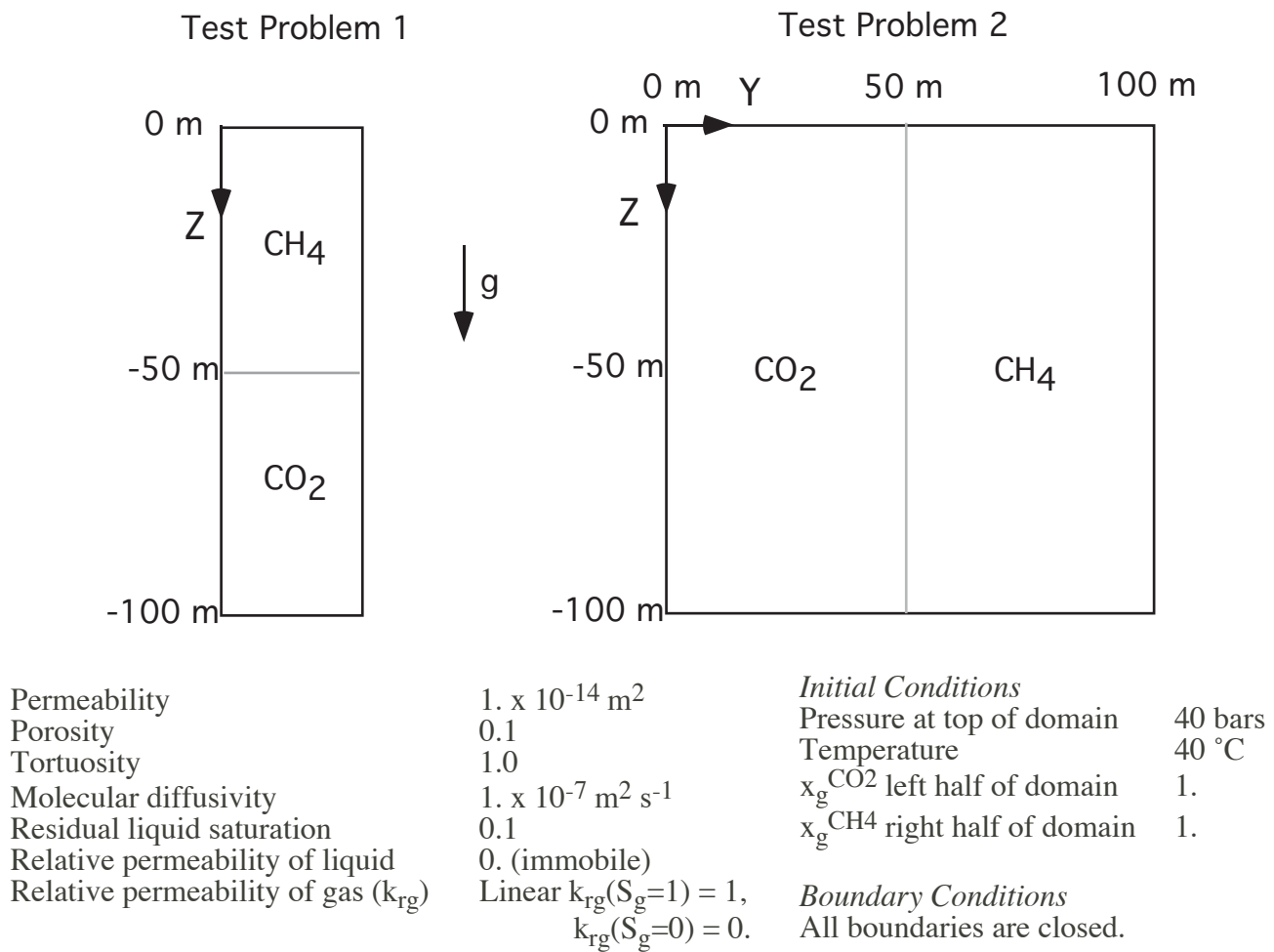
PROPERTIES OF CO<sub>2</sub>-CH<sub>4</sub> GAS MIXTURES AND AQUEOUS SOLUBILITY AT 40 BARS, 40 °C.

	gas phase				aqueous phase	
Simulation Code	x <sub>g</sub> <sup>CH4</sup>	x <sub>g</sub> <sup>CO2</sup>	ρ (kg m <sup>-3</sup> )	μ (Pa s)	x <sub>l</sub> <sup>CH4</sup>	x <sub>l</sub> <sup>CO2</sup>
CHEMTOUGH	0.	1.	105.39	1.49 x 10 <sup>-5</sup>	0.	1.64 x 10 <sup>-2</sup>
GEM	0.	1.	85.41	1.75 x 10 <sup>-5</sup>	0.	1.50 x 10 <sup>-2</sup>
SIMUSCOPP	0.	1.	85.35	1.02 x 10 <sup>-5</sup>	0.	1.24 x 10 <sup>-2</sup>
TOUGH2/EOS7C	0.	1.	85.45	1.70 x 10 <sup>-5</sup>	0.	1.62 x 10 <sup>-2</sup>
Reference Values	0.	1.	83.79 [3]	1.73 x 10 <sup>-5</sup> [3]	0.	1.37 x 10 <sup>-2</sup> [4]
CHEMTOUGH	0.5	0.5	46.88	1.34 x 10 <sup>-5</sup>	4.08 x 10 <sup>-4</sup>	7.45 x 10 <sup>-3</sup>
GEM	0.5	0.5	52.26	1.53 x 10 <sup>-5</sup>	3.82 x 10 <sup>-4</sup>	7.64 x 10 <sup>-3</sup>
SIMUSCOPP	0.5	0.5	52.29	1.11 x 10 <sup>-5</sup>	3.90 x 10 <sup>-4</sup>	6.20 x 10 <sup>-3</sup>
TOUGH2/EOS7C	0.5	0.5	51.97	1.44 x 10 <sup>-5</sup>	3.73 x 10 <sup>-4</sup>	8.07 x 10 <sup>-3</sup>
Reference Values	0.5	0.5	51.33 [3]	1.67 x 10 <sup>-5</sup> [3]	3.66 x 10 <sup>-4</sup> [5,6,7,8]	6.74 x 10 <sup>-3</sup> [5,6,7,8]
CHEMTOUGH	1.	0.	24.58	1.16 x 10 <sup>-5</sup>	7.49 x 10 <sup>-4</sup>	0.
GEM	1.	0.	26.48	1.22 x 10 <sup>-5</sup>	7.51 x 10 <sup>-4</sup>	0.
SIMUSCOPP	1.	0.	26.46	1.26 x 10 <sup>-5</sup>	7.81 x 10 <sup>-4</sup>	0.
TOUGH2/EOS7C	1.	0.	26.42	1.21 x 10 <sup>-5</sup>	7.43 x 10 <sup>-4</sup>	0.
Reference Values	1.	0.	26.10 [3]	1.23 x 10 <sup>-5</sup> [3]	7.22 x 10 <sup>-4</sup> [5,6,7,8]	0.

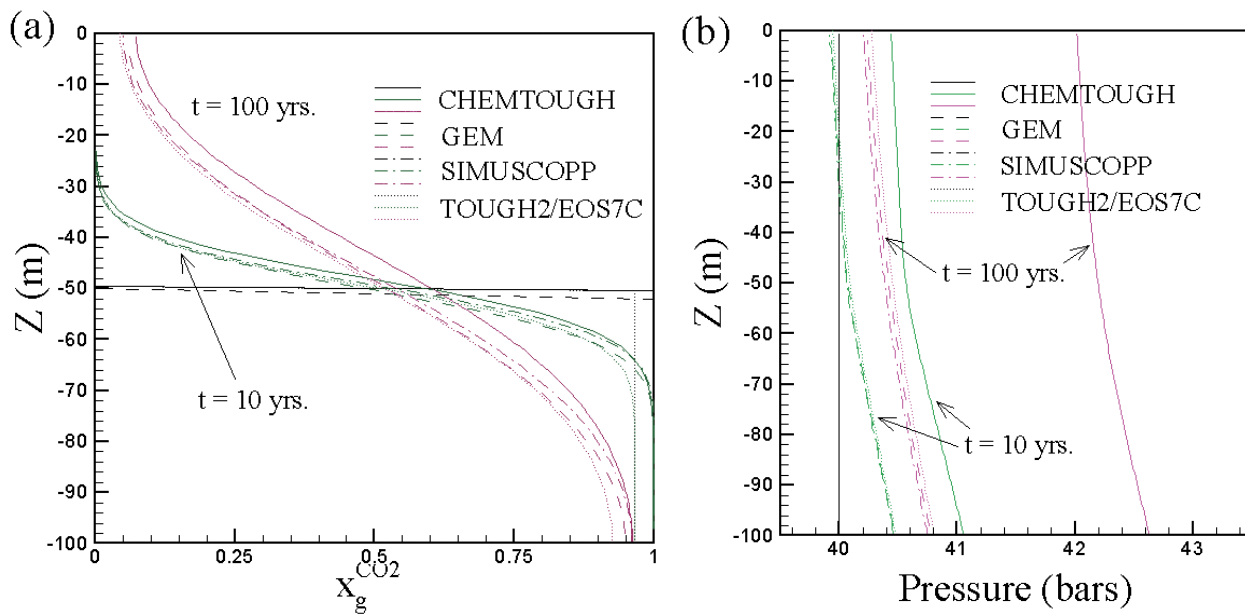
TABLE 2

PROPERTIES OF CO<sub>2</sub>-CH<sub>4</sub> GAS MIXTURES AND AQUEOUS SOLUBILITY AT 100 BARS, 40 °C.

	gas phase				aqueous phase	
Simulation Code	x <sub>g</sub> <sup>CH4</sup>	x <sub>g</sub> <sup>CO2</sup>	ρ (kg m <sup>-3</sup> )	μ (Pa s)	x <sub>l</sub> <sup>CH4</sup>	x <sub>l</sub> <sup>CO2</sup>
CHEMTOUGH	0.	1.	432.33	2.88 x 10 <sup>-5</sup>	0.	4.09 x 10 <sup>-2</sup>
GEM	0.	1.	564.82	4.35 x 10 <sup>-5</sup>	0.	2.39 x 10 <sup>-2</sup>
SIMUSCOPP	0.	1.	561.44	3.59 x 10 <sup>-5</sup>	0.	2.30 x 10 <sup>-2</sup>
TOUGH2/EOS7C	0.	1.	566.00	4.35 x 10 <sup>-5</sup>	0.	4.03 x 10 <sup>-2</sup>
Reference Values	0.	1.	631.90[3]	5.04 x 10 <sup>-5</sup> [3]	0.	2.19 x 10 <sup>-2</sup> [4]
CHEMTOUGH	0.5	0.5	130.58	1.41 x 10 <sup>-5</sup>	1.14 x 10 <sup>-3</sup>	1.61 x 10 <sup>-2</sup>
GEM	0.5	0.5	158.10	1.88 x 10 <sup>-5</sup>	8.27 x 10 <sup>-4</sup>	1.33 x 10 <sup>-2</sup>
SIMUSCOPP	0.5	0.5	158.44	1.46 x 10 <sup>-5</sup>	9.08 x 10 <sup>-4</sup>	1.15 x 10 <sup>-2</sup>
TOUGH2/EOS7C	0.5	0.5	155.16	1.81 x 10 <sup>-5</sup>	9.43 x 10 <sup>-4</sup>	2.00 x 10 <sup>-2</sup>
Reference Values	0.5	0.5	153.97 [3]	1.94 x 10 <sup>-5</sup> [3]	7.95 x 10 <sup>-4</sup> [5,6,7,8]	1.21 x 10 <sup>-2</sup> [5,6,7,8]
CHEMTOUGH	1.	0.	61.45	1.16 x 10 <sup>-5</sup>	1.87 x 10 <sup>-3</sup>	0.
GEM	1.	0.	71.78	1.39 x 10 <sup>-5</sup>	1.58 x 10 <sup>-3</sup>	0.
SIMUSCOPP	1.	0.	71.66	1.43 x 10 <sup>-5</sup>	1.82 x 10 <sup>-3</sup>	0.
TOUGH2/EOS7C	1.	0.	71.57	1.41 x 10 <sup>-5</sup>	1.86 x 10 <sup>-3</sup>	0.
Reference Values	1.	0.	70.03 [3]	1.41 x 10 <sup>-5</sup> [3]	1.54 x 10 <sup>-3</sup> [5,6,7,8]	0.



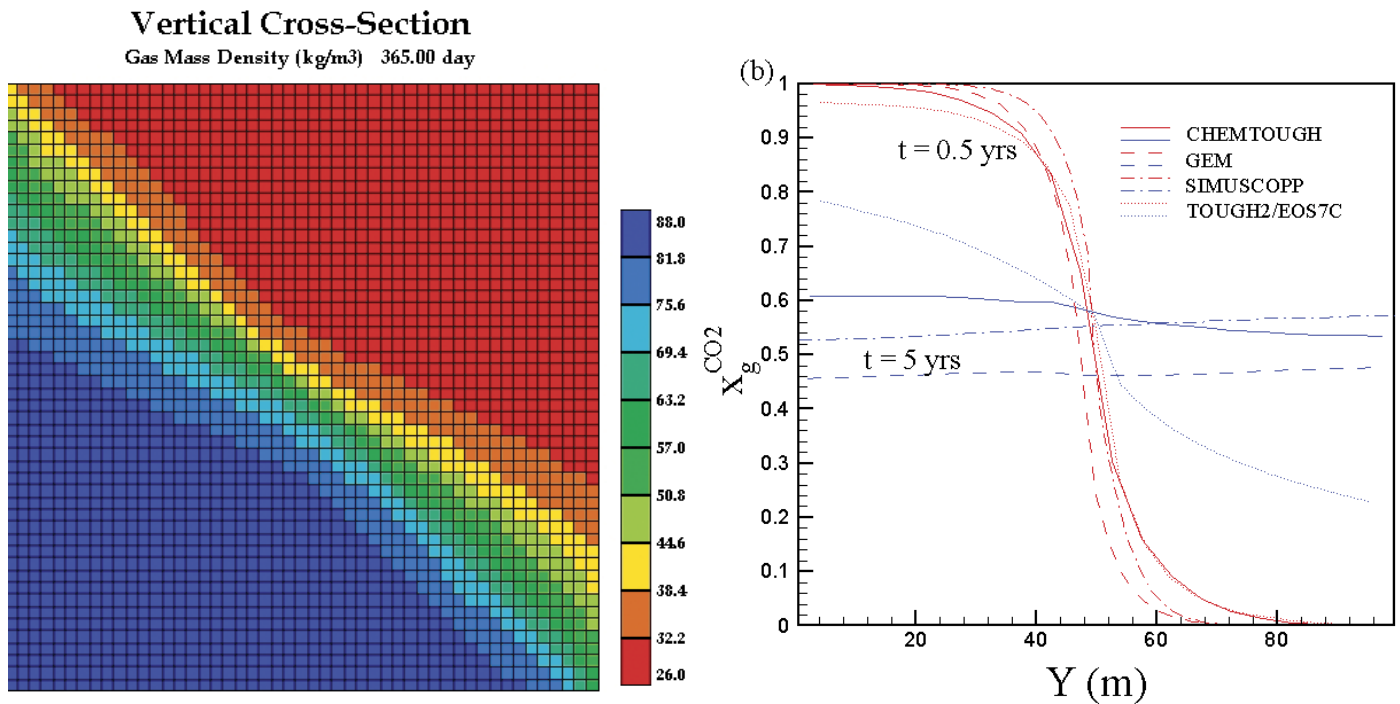
**Figure 1.** Specifications for Test Problems 1 and 2.



**Figure 2.** (a) Mole fraction  $\text{CO}_2$  in the gas and (b) pressure (bars) as a function of  $Z$  for Test Problem 1 at  $t = 0, 10$ , and 100 yrs.

## TEST PROBLEM 2. ADVECTIVE-DIFFUSIVE MIXING DUE TO LATERAL DENSITY GRADIENT

In this problem, CO<sub>2</sub> and CH<sub>4</sub> gases are placed side-by-side and allowed to mix. The strong lateral density gradient between the dense CO<sub>2</sub> gas and the relatively light CH<sub>4</sub> gas causes a strong density-driven flow where CO<sub>2</sub> tends to move downward and CH<sub>4</sub> tends to move upward to the top of the reservoir. Problem specification and domain schematic are presented in Figure 1. An example of the computed result at t = 1 yr is shown in Figure 3a by the density field as computed by GEM. Comparison of results is presented in Figure 3b as horizontal profiles of mole fraction CO<sub>2</sub> in the gas at two different times. As shown in Figure 3b, variations in results between the four codes are more pronounced than for Test Problem 1, showing that larger differences can be expected for cases of more complex flow and transport. However, concentrations along the profile at Z = -50 m are very sensitive to small variations in the simulation at late times since the interface between the gases is located in this region, and the profile effectively follows this interface.



**Figure 3.** (a) two-dimensional density (kg m<sup>-3</sup>) field at t = 1 yr. (b) horizontal profiles of  $x_g^{CO_2}$  at Z = -50 m.

## DISCUSSION AND CONCLUSIONS

Physical property estimates and simulation results for the mixing of CO<sub>2</sub> and CH<sub>4</sub> gases show fair to good agreement. Overall, the four simulation codes model the general processes of molecular diffusion and density-driven flow and advective mixing similarly, although results differ in detail. This comparison has brought to light differences in physical property estimates to which differences in simulated results are likely attributable. While process description and problem setup are subjective since they are influenced by the experience and approach of the practitioner, physical properties are objective and relatively well known. Code developers should endeavor to make physical property estimates more accurate. Nevertheless, in the actual practice of numerical simulation of subsurface processes, the largest differences between simulation results will likely be due to the conceptual models used, including assumptions about reservoir heterogeneity, as opposed to the algorithmic details of codes or physical property estimates.

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